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Atmospheric Measurement Techniques Discussions

Interactive comment on "Probing ice-nucleation processes on the molecular level using second harmonic generation spectroscopy" *by* A. Abdelmonem et al.

Anonymous Referee #2

Received and published: 29 June 2015

This is a very well-written paper presenting fundamental research on heterogeneous ice nucleation which is a very important topic related to cloud formation in the atmosphere. Sheet-like mineral surfaces (mica and sapphire) have been investigated focusing on the formation of ice-like structures, i.e. water structuring at the mineral surface, prior to the heterogeneous ice nucleation event. The authors apply second harmonic generation (SHG) in total internal reflection geometry (TIR). Up to my knowledge this is one of the first works, which really use this even-order nonlinear optical effect and make changes visible occurring in a water monolayer absorbed to a cold surface.

When reading the answers to referee H. Christenson, I learned that âĂŘ13 °C is high

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Interactive Discussion
Discussion Paper

and cannot be the heterogeneous freezing point of any of the selected surfaces. Therefore, I conclude that the ice nucleus, which is neither mica nor sapphire, triggers the ice nucleation independently and subsequently causes the ice formation also on the mineral surface (with a slight temperature difference of 2-3°C). That means that water is pre-structuring on mica but is not on sapphire at temperatures between 0 and -13°C. Both observations, however, are independent from the original ice nucleation event caused by something else. If my understanding is correct than you should rewrite the results and discussion section giving the focus more on the pre-ordering, since the signal drop, i.e. the ice coverage of the surfaces, is a secondary effect and is not your main focus.

You might also discuss the possibility if something else than a water monolayer could have caused the slight signal increase at the surface of mica. Can you exclude a temperature dependence of the optical parameter of mica? Is the temperature difference between both signal drops of 2-3°C really significant and is a pre-structuring of the water layer on mica the only interpretation for the earlier freezing on mica? I also wonder if the area enclosed by the vertical trend line of the laser signal during freezing and the curve alteration due to the latent heat is proportional to the mass of ice being formed. If we assume so, for what reasons should both areas be the same as in your experiment? From my point of view these questions are important in order to prove the statement: "...that heterogeneous ice crystallization on a surface is related to the degree of order induced by this surface to supercooled water".

Comment on the set-up: Please, explain the SHG method in more detail indicating which vibrational states are excited and due to which symmetry rules. What are the experimental prerequisites to allow SFG studies to gain more direct information on the interfacial water molecules?

Comment on figure 1: Your legend should also include the symbol of the lenses. The inset with the sample geometry should be turned upside down in order to make it comparable with the schematic presentation.

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The paper should be published as it is after some corrections of chapter 3 and 4.

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 5265, 2015.